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**ROLE OF GRADIENT AND
MULTISCALE INTERFACE
MORPHOLOGY IN THREE-
DIMENSIONAL REINFORCEMENTS
IN COMPOSITES (PREPRINT)**

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14. ABSTRACT The interface morphology tailoring between the constituent phases in composites (e.g., fiber-matrix, inter-lamina, inter-yarn, nano-constituents-matrix, etc.) is essential in optimizing composite properties. In the case of composite strength, the mismatch of properties between the phases causes stress concentration at the interfaces, which in turn causes the initiation of damage and failure. A way of minimizing the mismatch of properties at the interface is demonstrated to reduce the interface stress concentration and hence delay the damage initiation process together with improving composite strength. Further, controlling the interface impedance mismatch is also important in controlling scattering of thermal energy at the interface to tailor thermal conductivity of composites, especially in the thickness direction. A gradient interface material morphology is thus desirable to enhance strength as well as other properties (e.g., thermal) of composites. Incorporation of nano constituents in composites now potentially enable us to implement the gradient interface morphology at multiple scale level, from nano meter (nano constituent interface) to laminate ply interlayer (micro meter scale). In this study, the effect of implementing the gradient interface at different scale level is reviewed to assess its potential in enhancing composite strength and thermal properties.					
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Role of Gradient and Multiscale Interface Morphology in Three-Dimensional Reinforcements in Composites

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The interface morphology tailoring between the constituent phases in composites (e.g., fiber-matrix, inter-lamina, inter-yarn, nano-constituents-matrix, etc.) is essential in optimizing composite properties. In the case of composite strength, the mismatch of properties between the phases causes stress concentration [1, 2] at the interfaces, which in turn causes the initiation of damage and failure. A way of minimizing the mismatch of properties at the interface is demonstrated to reduce the interface stress concentration [2] and hence delay the damage initiation process together with improving composite strength. Further, controlling the interface impedance mismatch is also important in controlling scattering of thermal energy at the interface to tailor thermal conductivity of composites, especially in the thickness direction. A gradient interface material morphology is thus desirable to enhance strength as well as other properties (e.g., thermal) of composites. Incorporation of nano constituents in composites now potentially enable us to implement the gradient interface morphology at multiple scale level, from nano meter (nano constituent interface) to laminate ply interlayer (micro meter scale) [3, 4]. In this study, the effect of implementing the gradient interface at different scale level is reviewed to assess its potential in enhancing composite strength and thermal properties.

I. Introduction

It is a well known fact that the discontinuity of materials properties in heterogeneous materials systems causes stresses concentration (stress rise) at the vicinity of the interface between the two materials. One such example is the free-edge stress field in laminated composites, observed by Pagano, et. al [3-5]. The free-edge stress is the singular stress field at the materials interface at the free-edge of two adjacent laminae of dissimilar materials properties, arising due to different orientation of the laminae. The nature of the singular stress free-edge stress field, as predicted by Pagano [3, 4], is shown in Figure 1 below. The presence of such high stress concentration (of high stress gradient) causes the initiation of damage at the lamina interfaces [5]. The singular out-of-plane shear stresses at the interface of the intersection of three material juncture in textile composites (Figure 2), an analogous phenomenon of the free-edge stress field, is also observed by Roy, et. al [6, 7], as shown in Figure 3. It should be noted that the location of the three-material juncture in textile composite is not a single point; rather it is the line of a yarn intertwining with other yarn in presence of the matrix material. The presence of the two cases of interface stress singularity, as illustrated above, is essentially due to the mismatch of materials properties of the adjoining materials in the vicinity of the interface.

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There are many other materials configuration exists exhibiting the presence of interface stress concentration in different scale levels. For example, the fiber matrix interface stress concentration phenomena are the primary cause of the fiber-matrix damage initiation, limiting the strength of the material.

One possible approach of delaying the fiber-matrix or inter-yarn interface damage initiation resulting in enhancing strength of composite materials is to incorporate gradient materials morphology at appropriate scale level to minimize the extent of materials property mismatch at the vicinity of the interface. Such approach has been undertaken in the past, however, was essentially limited to layered materials property variation at the interface, which was not considered true gradient materials morphology. The advent of nano constituents of different shapes and sizes offers a renewed opportunity of engineering gradient materials morphology in multiple scale level.

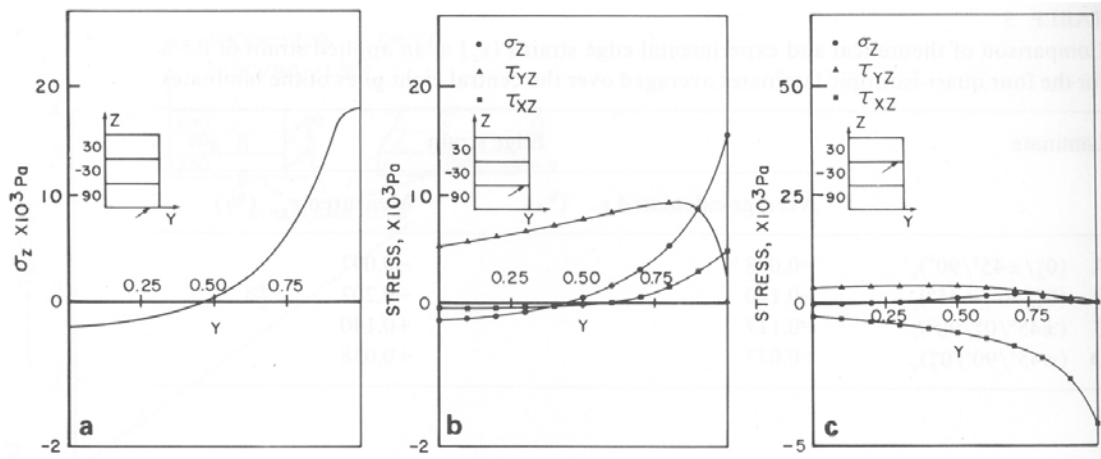


Figure 1. Interlaminar normal and shear stress distribution at ply interfaces of $[30/-30/90]_s$ laminates under uniaxial tension loading. (a) $[90/90]$, (b) $[-30/90]$ and (c) $[30/-30]$ interfaces; Reference [3].

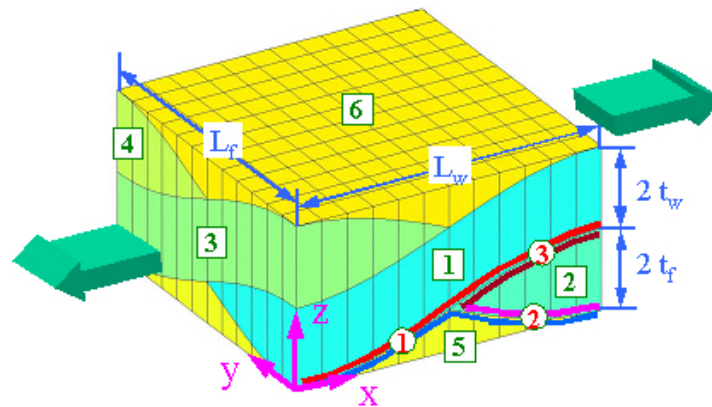


Figure 2. Schematic reinforcement configuration of plain weave textile composites showing yarn interfaces and location of three-material juncture (where lines 1, 2, and 3 intersecting).

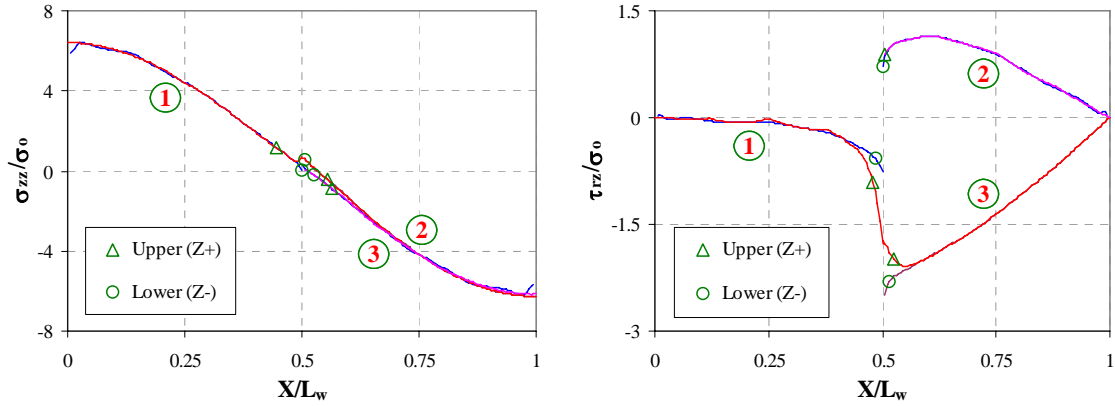


Figure 3. Interfacial normal ($\hat{\sigma}_3$) and shear stress ($\hat{\sigma}_5$) distributions on a unit cell of undamaged woven composites along three inter-yarn boundaries calculated with three different mesh schemes. Numbers in circles are interface numbers; Reference [6].

II. Gradient Interface Morphology Concepts

Although the gradient interface concept in reducing the extent of stress concentration factors at the interface region in composites is not new, the current innovative processing technology of controlled and aligned dispersion of nano constituents in composites provides a new opportunity of tailoring gradient materials morphology of composites interfaces. The advantages of the nano constituent sizes in the nanometer scale, the gradient interface morphology even at the fiber-matrix interface level (~ 10 nm thickness) is now close to reality. Also, nanofibers dispersed in thin layers (~ 100 - 150 nm), provides another gradient interface concept at the interface between laminae (plies) of laminated composites. Sihn, et. al [8, 9] demonstrated enhancement of delamination resistance in laminated composites by processing electrospun thin nanolayers (NL) at the interfaces of laminated composites. Thin polymer nanolayers were first produced by randomly depositing electrospun nanofibers in a mat of thickness about 100 - 150 nm and B-staged before co-curing in the composite laminates. To demonstrate the potential of using nanolayer in suppressing (delaying) the free-edge induced interface delamination, the laminate stacking sequence of $[+30/-30/90]_s$, which is known to induce severe free-edge stress field causing interlaminar delamination, was used as the base line laminate. The nanolayers (NL) were inserted at each lamina interfaces of the above laminate and processes to make the $[+30/NL/-30/NL/90/NL]_s$, with nanolayers. The interface property mismatch is expectedly reduced by incorporation of this nanofibers-reinforced nanolayer at the interface between the laminae. The stress level at the first microcracking damage, delamination damage and ultimate load drop increased by 8.4% , 8.1% and 9.8% , respectively (Figure 5b), with the addition of the nano-interlayers as compared with the pristine specimens. The number of microcracks decreases by 21.6% with the addition of the nano-interlayers (Figure 5a), which is not negligible. Thus, the electrospun nanofibers-reinforced nanolayer in laminate lamina (ply) interfaces demonstrates some success in reducing microcracking and delaying delamination by apparently reducing the

magnitude of interface stress concentration, which may be attributed to the reduction of the interface property mismatch by incorporating interface nanlayers.

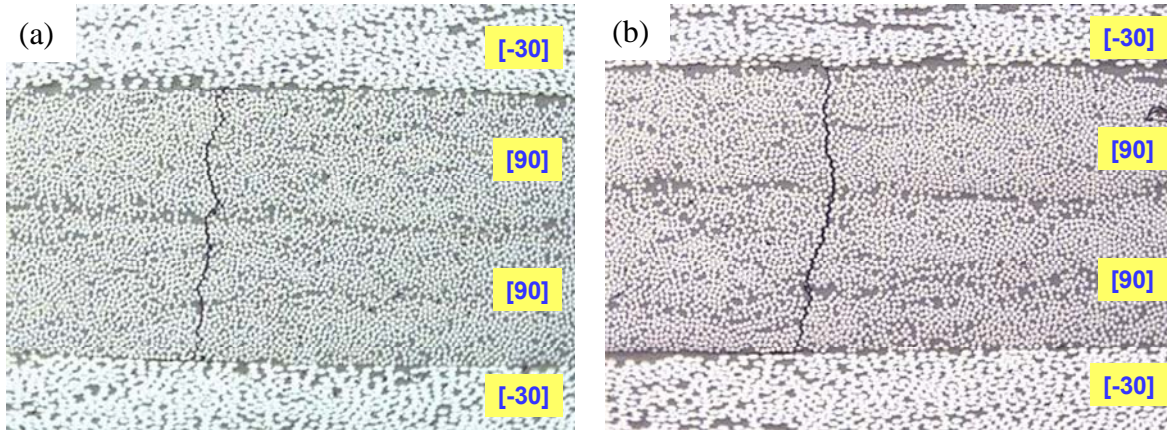


Figure 4. Micrographs with a microcrack at the first-ply failure under a uniaxial tension loading at a stress level of (a) 165.2 MPa (24.0 ksi) without nano-interlayer $[+30/-30/90]_s$ and (b) 171.2 MPa (24.8 ksi) with nano-interlayer $[+30/NL/-30/NL/90/NL]_s$, reference [7]

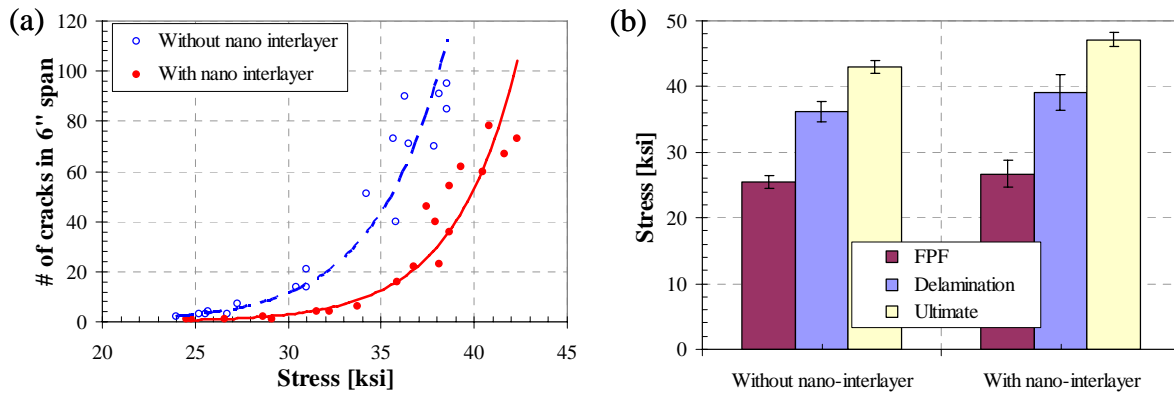


Figure 5. (a) Number of microcracks counted in a 6-inch span at the free edge in the middle of the test specimen at various stress levels and (b) stress levels at the first-ply failure, delamination and ultimate failure with and without the nano-interlayers under uniaxial tension loading; Reference [7].

The gradient interface morphology at the fiber-matrix interface may potentially be tailored to enhance composite strength. In-situ polymerization of matrix (thermoset resin) with proper surface functionalization of fibers may provide gradient interface properties. The in-situ polymerization of functionalized nanofiber interface in epoxy matrix shown to produce gradient epoxy modulus (qualitatively characterized by EELS, Figure 7) resulting in suppressing failure at the fiber-matrix interface, as shown in Figure 6 below. One of the challenges is in quantifying the gradient material modulus at the fiber-matrix interface (of thickness in the order of 20-30 nm), without which the benefit of interface gradient morphology can not truly be determined.

Besides in-situ polymerization, other approaches, such as, growing nanotubes on carbon fiber surface, on composite perform surfaces, are potentially attractive in implementing gradient morphology at the material interface, Figure 8 [11, 12].

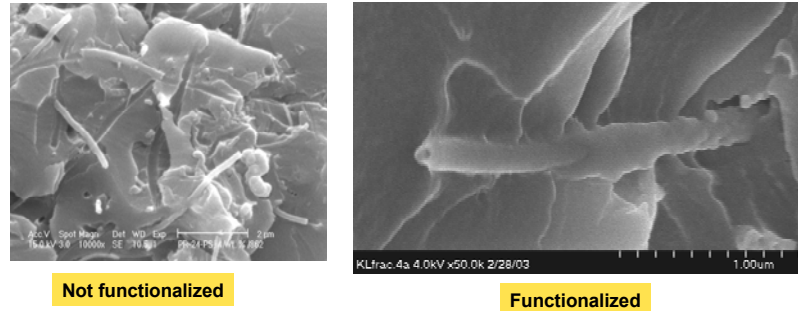


Figure 6. Micrographs of the failure surface of unfunctionalized and functionalized nanofibers in Epon 828. The strength for the case of functionalized fiber apparently increased by 140%; Reference [10].

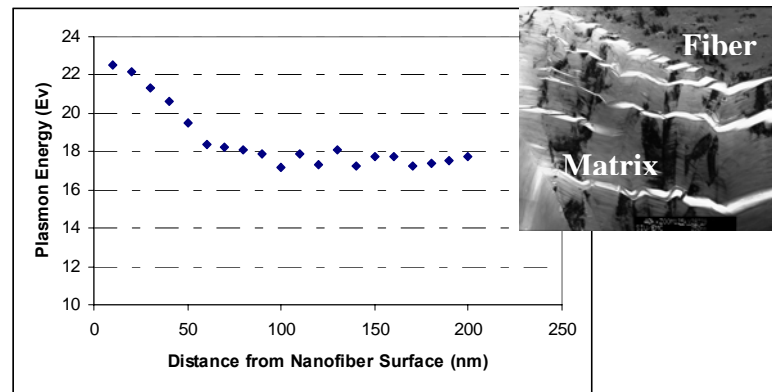


Figure 7. Qualitative characterization of fiber-matrix interface material property with Electron Emission Loss Spectroscopy (EELS) by correlating Plasmon energy with material modulus; Reference [10].

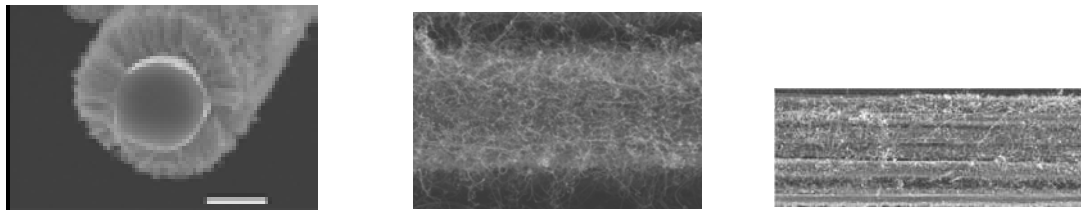


Figure 8. Carbon nanotubes grown on fiber and composite perform surfaces.

III. Stress Analysis of Gradient Interface

Along with developing processing techniques to incorporate gradient material morphology at composites interface, appropriate stress analysis needs to perform to quantify the gradient morphology effect on the failure process. Pan [2] studied the nature of interface stress field in presence of gradient material property at the interface. With

his linear elastic analysis he showed that tensile radial stress, which contributes to the cause of fiber-matrix cracking, can be suppressed with suitable interface gradient morphology, Figure 9.

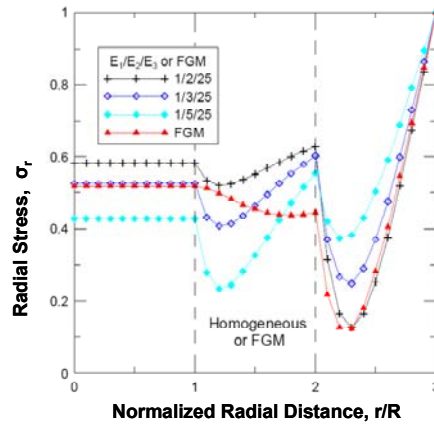


Figure 9. Distribution of radial stress at the fiber-matrix interface region of a concentric cylinder model of layered and gradient interface properties; Reference [2].

IV. Summary

The prospect of incorporating gradient materials morphology at different scale level of composite interface (fiber-matrix, inter-yarn, inter-lamina, etc.) in minimizing the materials property mismatch at the interface is reviewed. A few processing schemes of incorporating gradient interface concepts, including nano tailoring of fiber-matrix interface, is presented qualitatively illustrating their potential in delaying interface cracking (or delamination) phenomena. Appropriate characterization tools are needed to truly quantify the effect of gradient interface in enhancing composite performance (mechanical as well as thermal).

V. References

1. V. Buryachenko, A. Roy, K. Lafdi, K. Anderson, S. Sihn, S. Chellapilla, 2005, "Multi-scale mechanics of nanocomposites including interface: Experimental and numerical investigation," *Composites Science and Technology* (20th Anniversary Volume), Vol. 65, pp 2435-2465.
2. Pan, E. and Roy, A. K. (2006): A simple plane-strain solution for functionally graded multilayered isotropic cylinder. *Structural Engineering and Mechanics* (in press).
3. Pipes, R. B. and N. J. Pagano, 1970, "Interlaminar Stresses in Composite Laminates Under Uniform Axial Extension," *Journal of Composite Materials*, **4**, pp. 538-548.
4. Pagano, N. J. and S. R. Soni, 1983, "Global-Local Laminate Variational Model," *International Journal of Solids and Structures*, **19**, No. 3, p. 207.
5. Kim, R. Y. and S. R. Soni, 1984, "Experimental and Analytical Studies on the Onset of Delamination in Laminated Composites," *Journal of Composite Materials*, **18**.
6. A. K. Roy and S. Sihn, "Development of a Three-Dimensional Mixed Variational Model for Woven Composites: Part I – Mathematical Formulation," *Intl. Journal of Solids and Structures*, Vol. 38, 2001, pp 5935-5947.

7. S. Sihh, E. V. Iarve and A. K. Roy, 2003, "Three-Dimensional Stress Analysis of Textile Composites: Part I. Numerical Analysis," International Journal of Solids and Structures, Vol. 41, No. 5-6, pp. 1377-1393.
8. S. Sihh, J. W. Park, R. Y. Kim, W. Hah, A. K. Roy, Prediction of Delamination Resistance in Laminated Composites with Electrospun Nano-Interlayers using a Cohesive Zone Model, Paper Number : AIAA-2006-1771, AIAA SDM Conference, Newport, RI, 1-4 May 2006.
9. S. Sihh, J.W. Park, R.Y. Kim, and A.K. Roy, Improvement of Delamination Resistance in Composite Laminates with Nano-interlayers, SAMPE 2006, April 30 – May 4, Long Beach, CA, 2006.
10. Roy, A. K., Mechanics of Nanofiber Composites, 3rd US-Korea Workshop on Nanostructured Materials & Manomanufacturing, Hanyang University, Seoul, Korea, 10-11 May 2004
11. Chen, Q. D.; Dai, L. M., Plasma patterning of carbon nanotubes, Appl. Phys. Lett. 2000, 76, 2719-2721.
12. Huang, S. M.; Dai, L. M.; Mau, A. W. H., Controlled fabrication of aligned carbon nanotube patterns, Physica B 2002, 323, 333-335.